VARIABILITY, UNCERTAINTY, AND SENSITIVITY OF PHOSPHORUS DEPOSITION LOAD ESTIMATES IN SOUTH FLORIDA

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Abstract. Atmospheric deposition, a substantial source of phosphorus (P) to the Florida Everglades, has been measured on a weekly basis in South Florida since 1974, but P measurements are highly variable due to random noise in the data. This study applies statistical approaches that calculate the variability and uncertainty of the P load estimation model based on wet and dry P concentrations and rainfall volume. The average mean and standard deviation of the estimated P deposition rates for 13 sites in South Florida are 41±33 mg P m^{-2} yr^{-1}. First order analysis of the random and measurement errors in the input variables produces a propagation error estimate in P load calculation. The atmospheric P deposition load shows high spatial and temporal variability with no consistent long-term trends. Because of the random noisy nature of P deposition, estimated P deposition loads have a significant amount of uncertainty no matter what type of collection instrument is used. Thus, duplicate sampling is highly recommended to increase the amount of uncontaminated data.

Keywords: atmospheric deposition, first order analysis, monitoring design, sensitivity and error analyses, spatial variability, wet and dry phosphorus deposition

1. Introduction

Anthropogenic phosphorus (P) loads to the Everglades of South Florida have resulted in significant changes to this oligotrophic ecosystem (Davis, 1994). As a result, the State of Florida enacted a program to reduce phosphorus loading to the Everglades through a series of best management practices and constructed wetlands known as Stormwater Treatment Areas (State of Florida, 1994). To manage these phosphorus loads, accurate monitoring and analysis are required of both controllable and non-controllable sources. In South Florida, where most water bodies are large and shallow, atmospheric deposition, a non-controllable source, is a significant contributor of phosphorus (Chen and Fontaine, 1997). Atmospheric deposition will become even more significant, as controllable loads from agricultural regions are reduced.

Atmospheric deposition is commonly sampled in two separate forms: wet (rainfall) and dry (dustfall). The South Florida Water Management District (District) has collected atmospheric deposition data in the region since 1974. The monitoring program was significantly improved in 1992 with the deployment of wet/dry

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collectors (Aerochem Metrics Model 301 automatic wet/dry sampler) and adoption of a standard operating procedure for data collection and processing according to recommendations of the National Atmospheric Deposition Program (Bigelow and Dossett, 1988).

Many sources contribute to atmospheric P deposition. These include a combination of oceanic aerosols, dust from agricultural practices, burning, soil erosion, industrial and automobile pollution, etc. A primary concern with these potential sources of nutrient-bearing materials is their location of origin. If they originate inside the area of interest, then they may be viewed as local recycling or sources of contamination, such as frogs, bird droppings, and insects. If they originate outside the area of interest, such as some ash, dusts and vegetation debris and are transported by atmospheric processes, then they are a true part of the atmospheric deposition. It is almost impossible to determine the origin of P-bearing materials in routine monitoring. A secondary concern is the impact that these sources may have on P load estimates. If they add large amounts of P (such as bird droppings), then they will bias the estimate. If they add very little P (such as insect parts), then there is no contamination problem. The challenge in analyzing data from a monitoring network is to remove the bias while retaining the true signal of P depositions.

Another concern in deposition monitoring is the sampling of dry deposition. Techniques for estimating dry deposition include methods of: micro meteorology; surface accumulation; throughfall; watershed mass balance; and other inferential techniques (Erisman *et al.*, 1994). The District has used the surface accumulation method based on dry buckets to measure dry atmospheric deposition. The dry bucket method is simple, inexpensive, and, therefore, commonly used in field. In particular, this method is useful for measuring deposition of large particles (Hicks, 1986; Erisman *et al.*, 1994). Since P is primarily associated with particles greater than 2 μm in diameter (Graham and Duce, 1982; Lawson and Winchester, 1979), the dry bucket may be an adequate sampler of P dry deposition in this region (Ahn and James, 1999a).

The objectives of this study are to estimate the total weekly P load into South Florida from atmospheric deposition, and to define the variability and uncertainty in the P load estimates using a first order analysis.

2. Materials and Methods

2.1. Collection, Laboratory Analysis, and Pre-Processing of the Data

The District collects wet and dry deposition samples at weekly intervals from 18 monitoring sites and one replicate sampling site (BG2) (Figure 1). Each site has a set of Aerochem wet/dry buckets placed on a 1-m-high table. A movable lid operated by a moisture sensor plate is designed so that the lid moves over and covers

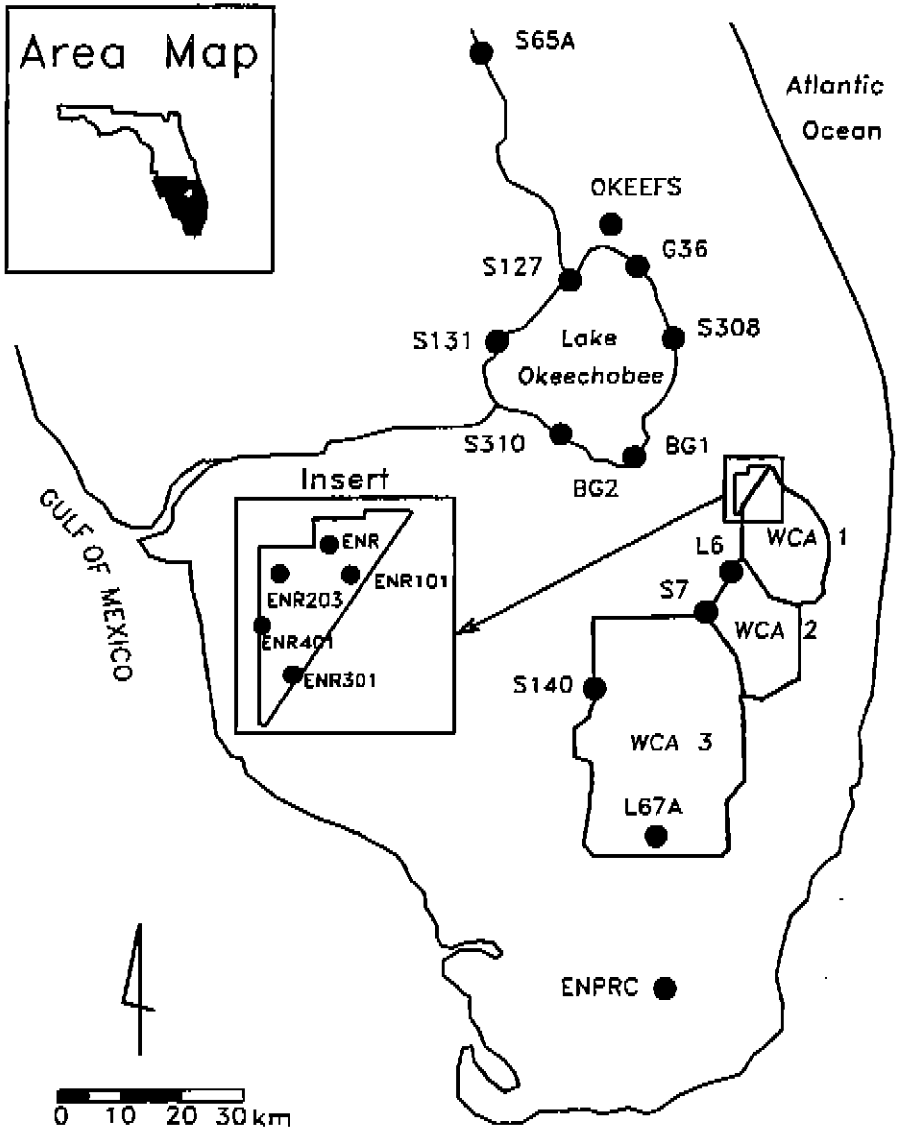

Figure 1. Location of atmospheric deposition monitoring sites operated by the District, (WCAs are water conservation areas and ENR is the Everglades nutrient removal project).

the dry bucket when it is raining, and covers the wet bucket when it is not raining, to prevent evaporation. The Aerochem bucket opening has an area of 0.0647 m^2 and a height of 0.25 m. This study selected wet and dry samples collected from April 7, 1992 to October 22, 1996. However, the actual record lengths vary from site to site owing to the periodic expansion of the monitoring program.

Before samples are collected, the wet and dry buckets are inspected in the field for contamination. Some types of contamination (e.g., insect and insect parts, amphibians, and reptiles) are removed with tweezers. Any visible contamination is identified according to 39 possible contamination sources and recorded in field notes. The buckets are sealed and transported to the laboratory in an upright position, preferably in a large cooler.

Wet samples are analyzed in the laboratory on the day after the field collection. The weight of rainfall in the wet bucket is measured using a Mettler Top Loading Balance (that is converted to rainfall depth) and an individual aliquot is taken from the bucket. For a dry bucket, one liter of deionized water is added into the bucket in order to make solution of the nutrients in the deposited materials and to rinse the sides of the bucket. The inside of the bucket is rubbed with a precleaned plastic spatula and an aliquot is then taken. Each water sample is placed into multiple 175 mL bottles and acidified with a 50% reagent grade solution of H_2SO_4 to a pH less than 2. Samples are digested with persulfate and P concentration is determined colorimetrically (USEPA, 1979). Quality assurance and quality control are performed in accordance with District standards (SFWMD, 1996).

In an effort to remove potential contamination, the samples were screened by an outlier detection approach. Potential outliers of both wet and dry P concentration data were identified first by field notes derived from visual inspection of the samples, and then by outlier detection statistics based on linear regression (Ahn and James, 1999a). As a result, 35% of wet samples and 18% of dry samples were removed from further data analyses. Then, the resulting data gaps in the P concentration data created by sample contamination and instrumental failures were filled by a statistical model which is based on a multivariate stochastic time series theory (Ahn and James, 1999b).

2.2. Spatial Variation, Sensitivity and Error

With the measured weekly wet and dry P concentrations (C_w and C_d) in μg L^{-1} and rainfall in cm week^{-1}, The model for P deposition load (L) in μg P m^{-2} d^{-1} is expressed as,

$$L_w + L_w = \frac{(1000C_w)\cdot(0.01R)}{7\text{ days}} + \frac{(1000C_d)\cdot V}{A\cdot 7\text{ days}} = 1.429C_wR + 2.2 \tag{1}$$

where L_d and L_w are the wet and dry loads, respectively, and A (= 0.0647 m^2) is the bucket opening area and V (= 0.001 m^3) is the volume of the water added to each dry bucket. For simplicity, we denote the above model as a functional form of $L = f(x)$, where $x = \{x_i, i = 1, 2, 3\}$ represents the set of independent variables $\{C_w, C_d, R\}$. We can further decompose $\{x_i\}$ as $\{x_{ij}, j = 1, \ldots, 19\}$ where j denotes j-th location where samples are collected. The spatial correlation structures of the input (or output) variables can be analyzed by spatial correlation coefficient, $\rho(d)$, and spatial error variance, $\sigma_e^2(d)$. For two concurrent values (x_{ij} and $x_{i,j'}$)

observed with a separation distance (d) where $j' \neq j$, the two spatial functions are estimated respectively by,

$$\rho(d) = \mathrm{cov}[x_{ij} x_{i,j'}]/(\sigma_{ij}\sigma_{i,j'}) \tag{2}$$

$$\sigma_e^2(d) = \mathrm{Var}[|x_{ij} - x_{i,j'}|/2] \tag{3}$$

where σ is the sample standard deviation of the designated site, $E[.]$ is the expected value, cov[.] is the covariance operator, and Var[.] is the variance operator. Variogram and spatial error variance are similar in nature but computation and application are quite different. The former is defined mainly in two-dimensional space at a given time, while the later is applied for space-time data. The reason for adopting the spatial error variance is that it is equvalent to the random error defined by replicate samples at a near zero distance in space.

Because the parameters (coefficients) in the model are fixed, the input variables in Equation (1) are subject to sensitivity and error analysis. Sensitivity, $S(x_i)$, of the L to the input x_i is defined by the partial derivative of the function as $S(x_i) = \partial f(x)/\partial x_i$. A relative sensitivity coefficient, $S_r(x_i)$ is defined as,

$$S_r(x_i) = \frac{\partial f(x)}{\partial x_i}\frac{\mu(x_i)}{\mu(L)} \tag{4}$$

where $\mu(.)$ denotes a mean. The relative sensitivity gives the percent change in output L for the change in each input variable x_i. This dimension-less quantity allows the ranking of input variables in terms of their sensitivities.

Errors in the P load model estimates can also be attributed to random errors in the input variables, $\{C_w, C_d, R\}$. The random error in this case originates from limited observations or a single measurement representing an areal value. The model estimation error, e, is defined by the non-zero difference of true value and the model estimates from a set of erroneous independent variables. If the model is unbiased, the values of e are normally distributed with a mean of zero and a finite error variance of σ_e^2.

Error in the P load model can be attributed partially to measurement errors; the error that exists in measurements in the laboratory. This was calculated from the District's 1996 quality control data, especially those of equipment standards which are free from short-scale variability. Four sets of equipment standards with P concentration levels of 30, 75, 300, and 1220 μg P L^{-2} (a total of 104 samples) were measured at the District's laboratory. The variation of the measured P values, which is an indicator of laboratory processing error, was then fitted by linear regression:

$$\sigma_{Em}(P) = 1.873 + 0.0176P \tag{5}$$

from which the measurement errors (in μg P L^{-1}) corresponding to the pooled mean values were calculated.

For an error analysis, it is assumed that the errors are statistically independent of the load estimations, are identically distributed, and uncorrelated to each other (Troutman, 1982). Because the measurement error is already inbedded in the random error, the pure random error without measurement error component, $\sigma_E^2(x_i)$, is obtained by:

$$\sigma_R^2(x_i) = \sigma_e^2(x_i, d = 0) - \sigma_{Em}^2(x_i) \tag{6}$$

The standardized error on the mean in percent, $E(x_i)$, can be computed (Rabinovich, 1993) by:

$$E(x_i) = 100\, t_q \frac{\sigma_e(x_i)}{\mu(x_i)} \tag{7}$$

where $\mu(x_i)$ (or $\mu(L)$ if it is output variable) is the sample mean, t_q is the q percent point of the Student's t distribution depending on the confidence level α and the degrees of freedom $\nu = n$-1 with n as the number of samples.

2.3. First Order Analysis

The propagation error in P load estimates caused by erroneous input variables and parameters is calculated by the first order analysis since the model is linear. First order analysis comes from a Taylor series expansion of the model about its expected input variables (Taylor and Kuyatt, 1994; Haan and Zhang, 1996). Given Equation (1) as a functional form of $L = f\ \{x_i, i = 1, 2, 3\}$, where x represents the set of independent variables $\{C_w, C_d, R\}$, the first order approximation of the output error (propagation error) variance, $\sigma_P^2(L)$, can be given as,

$$\sigma_P^2(L) = \sum_{i=1}^{3}[S(x_i)\times \mathrm{Var}(x_i)] \tag{8}$$

where the sensitivity to i-th input variable, $S(x_i)$, is written as,

$$S(x_i) = \frac{\partial f(x|x = \mu(x_{i'}), i' = 1, 2, 3, i' = \neq i)}{\partial x_i} \quad \text{for} \quad i = 1, 2, 3. \tag{9}$$

Furthermore, the relative sensitivity, S_r, is given by,

$$S_r(y_i) = S(x_i)\frac{\mu(x_i)}{\mu(L)}, \quad \text{for} \quad i = 1, 2, 3. \tag{10}$$

Equation (7) is used to compute propagated error from erroneous input components. In addition, the fractional error from i-th input component is estimated by,

$$f(x_i) = \frac{S(x_i)\cdot \mathrm{Var}[x_i]}{\mathrm{Var}[L]}, \quad \text{for} \quad i = 1, 2, 3. \tag{11}$$

TABLE I

Summary statistics (time-averaged) for atmospheric P deposition data measured in South Florida from April 1992 to December 1996

Site	No. of data	P conc. (μg P L^{-1})		Rain (cm wk^{-1})	P load (μg P m^{-2} d^{-1})			
		Wet	Dry		Mean	S.D.	Skew	L_d/L_w [a]
BG1	166	8.3	37.8	2.77	109.2	93.0	1.84	3.3
BG2	166	11.0	37.1	2.77	114.0	99.0	1.66	2.6
ENPRC	166	7.7	30.2	3.00	89.6	110.8	3.09	2.9
ENR	240	10.0	40.0	3.20	134.7	106.5	2.03	1.9
G36	51	16.3	60.7	1.83	146.1	113.1	1.57	11.1
L67A	51	5.5	6.5	2.06	31.3	32.8	2.85	0.9
L6	51	7.8	29.0	2.54	93.1	60.7	1.15	2.2
OKEEFS	240	6.8	32.6	2.79	95.4	74.2	2.10	3.1
S131	166	10.8	36.5	2.29	109.0	89.1	2.12	2.8
S140	240	8.0	30.6	3.05	97.7	76.2	1.93	2.3
S310	166	9.3	40.3	2.24	113.5	83.7	2.05	3.6
S65A	240	13.1	69.8	2.87	209.5	147.8	3.14	2.8
S7	240	8.0	32.8	3.18	107.0	95.3	2.25	2.1
Average	168	9.4	37.2	2.66	111.5	90.9	2.14	2.8
Pooled mean	–	9.4	38.4	2.80	118.6	105.1	2.70	2.5

[a] The ratio of total dry to wet loads.

The assumption underlying the first order analysis is that the derivative terms higher than the first order term are not significant, which is the case of the L load model.

3. Results and Discussion

3.1. P DEPOSITION LOAD CALCULATION

This study analyzed the data from only 13 sites because the data from the remaining six sites (S127, S308, four Everglades Nutrient Removal sites) are highly contaminated. The weekly P deposition rates at each site were computed by Equation (1). Summary statistics for the P loads and the means of input variables in each site were then computed (Table I).

The average of all site means is 112 μg P m^{-2} d^{-1}; individual site means range from 31 μg P m^{-2} d^{-1} at a remote station in a marsh area of the Everglades (L67A) to 210 μg P m^{-2} d^{-1} at S65A, a site surrounded by improved pasture (Figure 1).

In addition to a high variability of site means (28~180% from the average), the standard deviation of samples at each site is very high, almost equivalent to the mean (the average coefficient of variation is 0.82). The pooled mean and standard deviation are 6 and 16% higher, respectively, than the corresponding site average values. Hicks *et al.* (1993) reported that estimates of atmospheric P value from dry deposition range from 4 to 10 times that of wet deposition. The ratio of dry to wet deposition loads in our data is about 3 while that of the concentrations is about 4 with a range from 1 to 11.

An important issue in atmospheric deposition in this region is whether the sources of P are local, regional, global, or some combination of these atmospheric sources. The high spatial variability in P loads (Table I) suggests that P is more likely from local sources affected by proximal conditions at sampling sites. Furthermore, the minimum P load observed at L67A site (31.3 μg P m^{-2} d^{-1}), which is a remote marsh area and presumably is less influenced by local pollutants, supports this hypothesis. We infer from this minimum value that the portion of P loading attributed from regional and global sources is less than about 31 μg P m^{-2} d^{-1} or 28% of the estimated average value (Table I). However, further research is needed to identify the P sources.

The estimate of yearly P load (40.7$\pm$33.2 mg P m^{-2} yr^{-1}) is consistent with estimates of P deposition from peat accretion data (35.5 mg P m^{-2} yr^{-1}; Walker, 1993) and from bulk collectors throughout Florida (50 mg P m^{-2} yr^{-1}; Hendry *et al.*, 1981). However, it is less than the value observed in the Tampa area from seven bulk collectors (93.3 mg P m^{-2} yr^{-1}; Dixon *et al.*, 1996). These comparisons provide a certain level of confidence regarding the District's sampling network and procedures that we have taken.

3.2. Variabilities and Trends

Time series of the monthly average P deposition rates of three arbitrary selected sites show no temporal trend in the data as evidenced by the slopes of the regression lines that are not significantly different from zero (Figure 2). A 6-month moving average used to approximate seasonal trends simply fluctuates due to abnormal high P rates that appeared randomly in time. The other sites have similar temporal patterns but are not presented.

To investigate the seasonality in the data, the P deposition loads from all 13 sites were pooled and the distributions of P loads by month of the year were plotted (Figure 3). The mean P values were lowest in January (86 μg P m^{-2} d^{-1}) and highest in October (148 μg P m^{-2} d^{-1}). The average of P loads during the wet season (June to October) is about 26% larger than that of the dry season. This is caused mainly by the seasonal rainfall pattern rather than the weak seasonal change in P concentration. A similar seasonal pattern to the means is also observed for standard deviations.

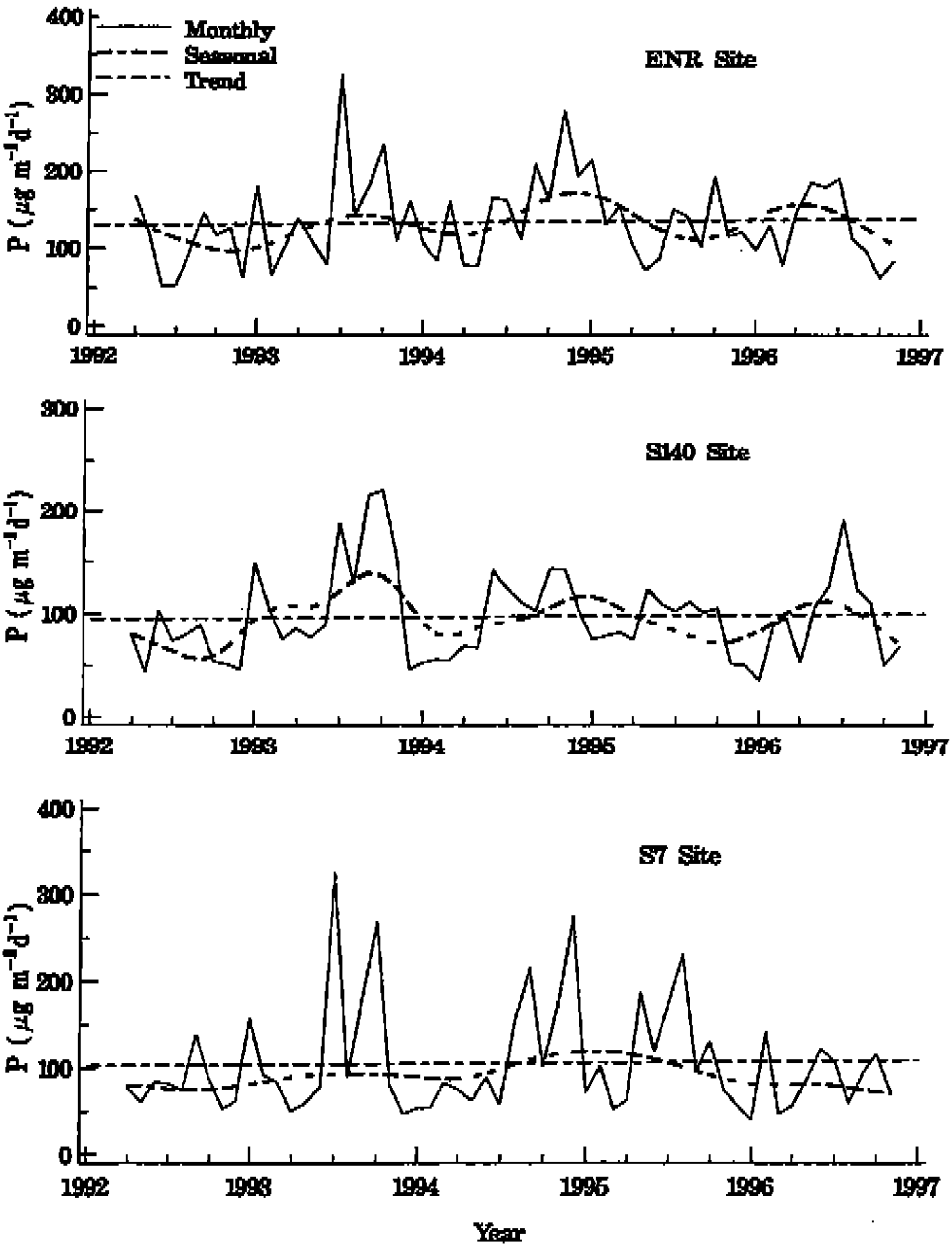

Figure 2. Seasonal and long-term trends for the phosphorus atmospheric deposition rates at three sites in South Florida.

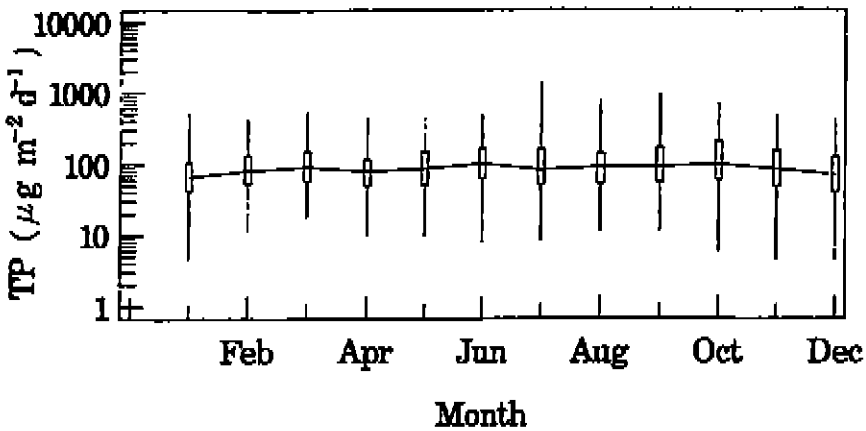

Figure 3. Box and whisker plots of P deposition rates from 13 sites in South Florida. The solid line represents the mean of all weekly P rates while the middle, bottom and top edges of each box are the median, 25th and 75th percentiles, and the bottom and top of whiskers are the minimum and maximum values, respectively.

The spatial correlation of rainfall decreases and the spatial variability of rainfall increases consistently with increasing separation distance between stations (Figures 4a, b, respectively). The spatial error variances of P load show very weak dependence to a separation distance (Figure 4e). The estimated spatial error variances are quite scattered indicating high spatial variability. The spatial error variances at zero distance are not zero. Instead, they are quite significant as the result of random noise components in the data.

3.3. Sensitivity and Error Analyses

Measurement errors, calculated from Equation (5), were 4 μg P L^{-1} and 6.25 μg P L^{-1} for the wet and dry samples respectively (Row 3, Table II). The measurement error in rainfall is the accuracy of rainfall data (about 3% of the measured rainfall depth). Random errors were computed using duplicate samples at BG1 and BG2 which are located about 3 m apart from each other. That is, for a set of duplicate samples (x_{i1} and x_{i2}) for i-th input variable at sites 1 and 2, there exists a non-zero difference between the two measurements. The true value was assumed the average of the two measurements. The errors in measured values are then obtained as $e = |x_{i1} - x_{i2}|/2$, from which the variances of the errors in P concentrations and load were computed (Row 4, Table II). These random errors are identical to the spatial error variances at zero separation distance (Figures 4b, c, d, f). The pure random error without laboratory processing error in each input variable can be computed by Equation (6).

First order analysis (Equations (8) through (11) indicates that the propagated error in P load estimates is 1923 μg P m^{-2} d^{-1} (Row 5, Column 4, Table II) which is about 25% of the mean P load estimate. The propagation error in P load estimate

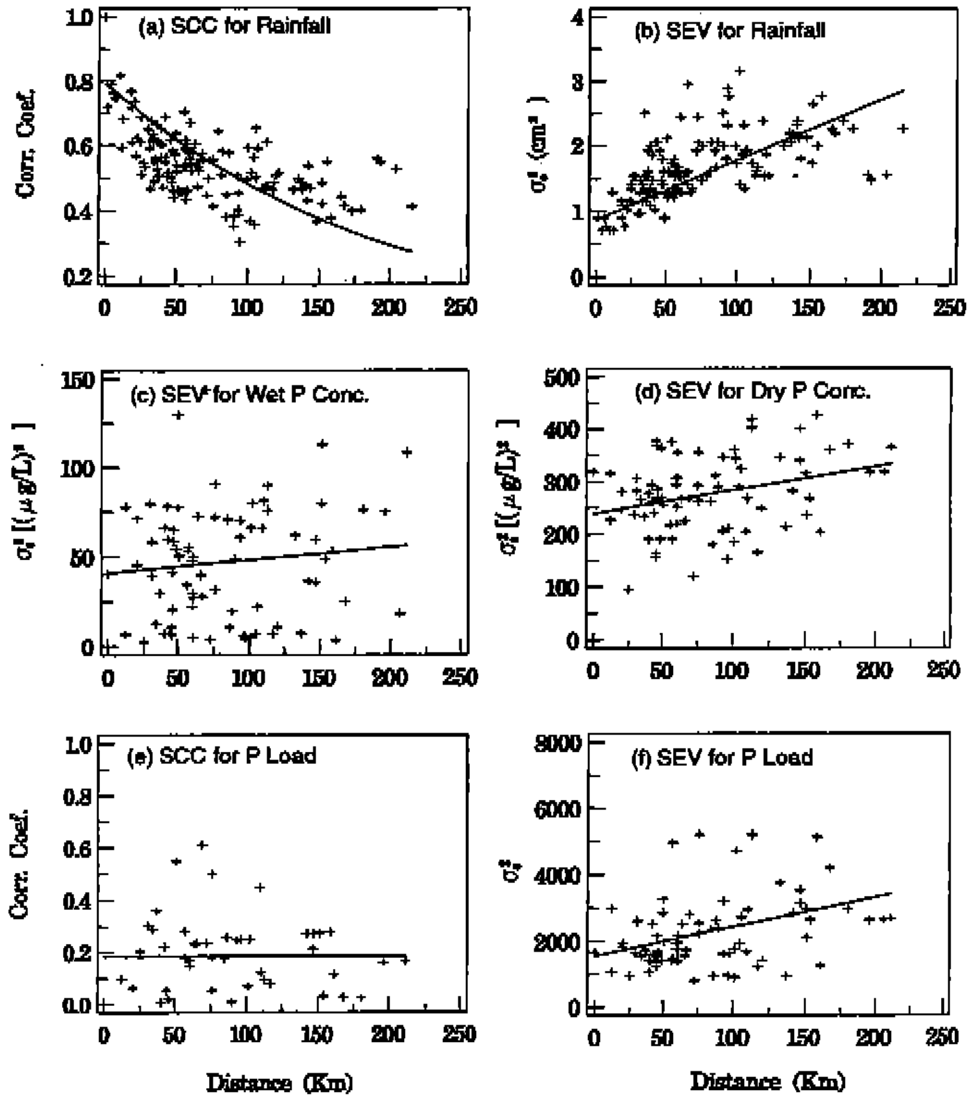

Figure 4. Spatial correlation coefficient (SCC) and spatial error variance (SEV) of the input and output components in the P load calculation, where the solid line in each plot is the line of best fit.

is greater than that of random error in P loads computed from the replicate data, 1444 μg P m^{-2} d^{-1} (Row 4, Column 4, Table II), demonstrating that first order analysis detects the combined propagation errors. The fraction of propagated error from the wet P concentration is much bigger than that of rainfall (Row 5, Table II). The majority of the error in the atmospheric loads are from dry P concentration measurement (57%), followed by wet P concentration (33%) and rainfall (9%). The relative sensitivities of wet P concentration and rainfall are equal and when

TABLE II

Result of sensitivity and error analyses

Statistics	Wet P conc., C_w (μg P L^{-1})	Dry P conc., C_d (μg P L^{-1})	Rainfall, R (cm wk^{-1})	P Load, L (μg P m^{-2} d^{-1})
(1) Mean, $\mu(.)$	9.39	38.44	2.80	118.56
(2) Variance, $\sigma^2(.)$	125.28	1358.59	12.27	11043.85
(3) Variance of measurement error, σ^2_{Em}	4.00	6.25	0.084	–
(4) Variance of random error, $\sigma^2_{\varepsilon}(0)$	35.62	222.50	0.872	1444
(5) Propagation error, σ^2_P, (or fraction on it)	(0.331)	(0.570)	(0.089)	1923.01
(6) Standardized error[a], $E(x_i)$, (%)	45.7	26.5	23.5	24.9
(7) Sensitivity, S	4.006	2.208	13.412	–
(8) Relative sensitivity, S_r	0.307	0.693	0.307	1.307 (sum)

[a] At 75% probability level.

added together equal the relative sensitivity of the dry (C_d) component (Row 8, Table II).

Also computed are the correlation coefficients of input and output variables, both in weekly concentration and P load terms (Table III). The P load is most influenced by dry P load or dry P concentration, followed by wet P concentration and rainfall. The correlation coefficients between load and each input variable (Row 6, Table III) are proportional to the corresponding fractional errors (Row 5, Table II) rather than the relative sensitivities (Row 8, Table II). The correlation between wet and dry P concentration is very low, indicating that they are two independent processes and one cannot be predicted by the other. The correlation between rainfall and P concentration is negative but very low, indicating that the dilution effect of large rainfall events in conjunction with simple scavanging models is not found in our weekly deposition data.

4. Summary and Conclusions

This study investigated the variability, sensitivity, and uncertainty in the P deposition load estimates in South Florida. From the results of this study, the following conclusions were drawn:

TABLE III

Correlation Coefficient Matrix for atmospheric deposition variables ($n = 2183$)

	Wet P conc. (C_w, μg P L^{-1})	Rain (R, cm wk^{-1})	Dry P conc. (C_d, μg P L^{-1})	Wet P load (L_w, μg P m^{-2} d^{-1})	Dry P load (L_d, μg P m^{-2} d^{-1})	P load (L, μg P m^{-2} d^{-1})
Cw	1.0					
R	–0.070	1.0				
Cd	0.116	–0.062	1.0			
Lw	0.463	0.561	0.025	1.0		
Ld	0.116	–0.062	1.0	0.025	1.0	
L	0.374	0.296	0.790	0.633	0.790	1.0

1. The estimated annual average P deposition rate in South Florida is about 41 mg P m^{-2} yr^{-1} with a standard deviation of 33 mg P m^{-2} yr^{-1}. The estimation error of the mean is about 25%. This error comes from random and measurement errors in both P concentration and rainfall. The ratio of dry to wet P loads is about three while that of P concentration is about four.
2. The means of P deposition rates vary from site to site ranging from 28 to 180% of the overall mean. The temporal variability of the P deposition loads is quite noticeable but the pattern is very irregular due to abnormal high P rates that appear randomly in time. The average P deposition during the wet seasons (June–October) is about 26% larger than that of the dry season. No long-term trend was found in the data.
3. Based on a first order analysis, the estimate of P loads is most sensitive to the dry P concentration measurement, followed equally by wet P concentration and rainfall. The same order was found in the fractional errors with different ratios. Unlike the sensitivity results, the propagation error in P load estimates caused by rainfall error is almost negligible.
4. Because of the random noisy nature of P deposition, estimated P deposition loads have a significant amount of uncertainty no matter what type of collection instrument is used. Duplicate sampling is highly recommended to increase the number of uncontaminated data. Identifying the source of deposition materials should be clarified through further research. Our agency has begun a project to investigate alternative sampling methods to identify any systematic instrumental error introduced by bucket samplers. The main purposes of this project are to reduce uncertainty in P load estimates and to develop a reliable cost-effective sampling method for atmospheric P deposition. It is hoped that this project will improve our models and reduce the uncertainty of atmospheric P load estimates in south Florida.

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